

AD-A065 676 ROCKWELL INTERNATIONAL ANAHEIM CA ELECTRONICS RESEAR--ETC F/6 8/7
EPITAXIAL GARNETS AND HEXAGONAL FERRITES, (U)
FEB 79 H L GLASS F44620-75-C-0045
UNCLASSIFIED C79-127/501 AFOSR-TR-79-0195 NL

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6 EPITAXIAL GARNETS AND HEXAGONAL FERRITES.

15 AFOSR CONTRACT F44620-75-C-0045

Final Scientific Report

10 Howard L. Glass Principal Investigator

11/13 Feb. 1979

9 Final rept. 1 Jan 75 - 31 Dec 78

I. RESEARCH OBJECTIVES

12 7p.
The overall objective of this research was to prepare single-crystal films of new and improved ferrite materials. The emphasis was on materials which will be useful in microwave and millimeter-wave signal processing devices. The research was concentrated on two classes of ferrites: the magnetic garnets typified by YIG ($\text{Y}_3\text{Fe}_5\text{O}_{12}$) and the hexagonal ferrites based on barium hexaferrite ($\text{BaFe}_{12}\text{O}_{19}$). In addition, some work was done on spinel ferrites including lithium ferrite ($\text{Li}_0.5\text{Fe}_{2.5}\text{O}_4$). Single crystal films of these ferrites were grown on appropriate substrates by the LPE (liquid phase epitaxy) process.

For the garnets, the basic LPE process had been well developed prior to the start of the contract. Therefore, research on these ferrites was directed toward improving their quality and suitability for microwave applications. Specific objectives for the garnets were: to minimize microwave losses (linewidth); to improve temperature stability of microwave resonance frequencies; to prepare and evaluate special layered structures by which the unique capabilities of the epitaxial film format can be exploited in microwave devices.

For the other classes of ferrites, LPE growth processes did not exist prior to the start of the contract. Therefore, the objectives were: to identify suitable substrates; to devise and test LPE processes; to evaluate the resulting materials. The hexagonal ferrites were chosen as target materials because their high magnetic anisotropies are advantageous in devices that operate at higher microwave and at millimeter-wave frequencies.

II. SIGNIFICANT ACCOMPLISHMENTS

We were fortunate in having many significant accomplishments during the four year contract period. The following accomplishments are most noteworthy.

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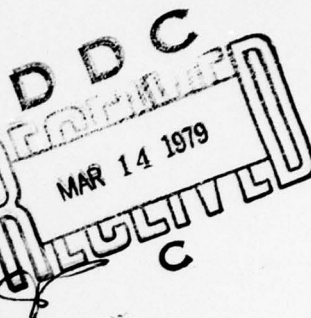
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1. Attainment of intrinsic linewidth in LPE YIG -

We showed that microwave losses in LPE YIG can be as low as losses in the best single-crystals grown by other methods. In fact, FMR (ferromagnetic resonance) measurements showed linewidths as low as the theoretical value for pure, perfect YIG crystals. The key to achieving the intrinsic linewidth lay in understanding the role of Pb as a constituent of the LPE flux and as an impurity incorporated into the YIG layer. In actual devices, attainment of intrinsic linewidth depends upon the ability to isolate single resonance modes; this is a function of device design.

2. Demonstration of temperature stability of the resonance frequency -

By recognizing that temperature drift in thin film samples is due not only to the direct temperature dependence of magnetization but to temperature variations of the several contributions to anisotropy as well, we were able to use controlled substitutions to bring the various temperature drifts into balance. Using combined La, Ga substitution in YIG, we stabilized the perpendicular resonance frequency. In more general terms, we learned to control the temperature drift so that, for example, it could be matched to the drift of a particular bias magnet. The technique should be applicable to other ferrites when they are in the form of single crystal films. There is a need for additional research and development to optimize the linewidth, which increases as a result of the substitution. Useful combinations of low linewidth and low temperature drift appear to be obtainable.

3. LPE growth of hexagonal ferrites -

We demonstrated that these ferrites can be grown by an LPE method analogous to that used for garnets. We grew three hexagonal ferrite types (known as M, Y and W) which differ in magnetic properties and, therefore, in potential device applicability. In addition to a kind of homoepitaxy on hexagonal ferrite substrates, we grew these ferrites on two kinds of non-magnetic spinel substrates: ZnGa_2O_4 and $\text{Mg}(\text{In,Ga})_2\text{O}_4$. We found that the hexagonal ferrite formed island or scale-like deposits, rather than uniform layers, on these spinel substrates. We still do not know whether uniform layers can be achieved. We discovered that ZnO , as a substrate constituent or as an LPE melt component, facilitates nucleation of hexagonal ferrite on spinel. A better understanding of this chemical effect along with improved substrate quality may permit us to grow uniform layers. In any event, the island deposits seem to be useful for some millimeter-wave devices and for studies of fundamental properties of hexagonal ferrites. Using island deposits, we have observed 35 GHz linewidths as low as 26 Oe, a value which is similar to the best reported bulk crystal values.

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4. LPE growth of lithium ferrite -

We found that the LPE method could be adapted to grow lithium ferrite on our spinel substrates. This ferrite has several potential advantages over YIG for certain kinds of microwave signal processing devices. The ability to grow good quality epitaxial layers of this material may permit realization of this potential.

5. Double YIG layer magnetostatic delay line -

We learned how to prepare multilayer composites in which a YIG film was grown on one side of a substrate wafer and another YIG film of different composition (and, therefore, having different magnetic properties) was grown on the other side. When such a composite is placed in a uniform magnetic field, the two YIG layers are biased to different frequencies. In this way, we obtained a linearly dispersive delay line having a time-bandwidth product an order of magnitude greater than conventional single YIG layer devices. Our composite represented an improvement over an earlier device (constructed in our laboratory by W. L. Bongiani under Air Force Contract F33615-72-C-1760) which employed two identical YIG layers in a specially constructed bias magnet that provided a field gradient. Our device, which employs a uniform field, is vastly simpler to build.

There were several other accomplishments.

We were able to grow very thick ($\sim 100\mu\text{m}$) YIG layers without significant reduction in quality. This result surprised many people since the LPE process has an inherent instability which, it was widely believed, invariably led to severe degradation when thick film growth was attempted. Thick YIG films have potential application in higher power and lower frequency microwave devices.

We obtained high uniaxial growth-induced anisotropies in YIG by incorporation of Pb. This was one of the first examples of high growth-induced anisotropy in the absence of magnetic rare-earths (which would introduce high losses). This effect led to a re-evaluation of the theory of growth-induced anisotropy.

We confirmed the report that Ru substitution in YIG can reverse the sign of the magnetostriction coefficient. We tried to use this effect to obtain temperature stability under compressive misfit stress; but the losses which accompanied Ru incorporation were excessive.

We demonstrated that substitution could be used to adjust the magnetic properties of LPE hexagonal ferrites just as is done in LPE garnets. Zn, Ti paired substitutions were made. By means of substitutions we can optimize the material for use at various frequencies.

We obtained a great deal of data on the effects of various growth parameters on LPE of hexagonal ferrites. We found that hexagonal ferrite type depends on growth temperature as well as on LPE melt composition. We measured the difference between saturation temperatures for deposition on spinel substrates and deposition on hexagonal ferrite substrates. This difference is a measure of the relative ease of nucleation. It provides a reference for studying the chemical effects which facilitate nucleation.

We have made substantial progress in analyzing magnetostatic wave propagation in multilayer structures containing two different ferrite layers. As indicated in paragraph 5, we can expect improved device performance from such materials.

We have grown YIG layers having gradients in their magnetic parameters and we have shown that these materials provide increased time-bandwidth products. However, these materials are difficult to analyze and further research is needed to determine their ultimate usefulness.

III. PERSONNEL

H. L. Glass served as principal investigator throughout the contract period. He carried out much of the crystal growth and crystallographic characterization. M. T. Elliott was a major contributor during the first two years of the contract and served as co-principal investigator during the second year. He performed the FMR analyses and other magnetic and microwave measurements. Dr. Elliott then transferred to another group in our laboratory, though he continued to work with us on an informal basis. For the third and most of the fourth year, microwave and magnetic characterization were taken over by J. H. W. Liaw. A few months before the contract period ended, Dr. Liaw transferred into a magnetic bubble group in another division, and was replaced by L. R. Adkins.

F. S. Stearns carried out many of the hexagonal ferrite growth experiments early in the contract period. Later, he devoted a substantial amount of his time to an in-house program to grow substrate crystals suitable for hexagonal ferrite LPE. W. L. Bongianini provided important guidance on microwave and millimeter-wave device requirements. He also performed many measurements on our materials.

IV. LIST OF PUBLICATIONS RESULTING FROM AFOSR SPONSORED WORK

1. F. S. Stearns and H. L. Glass, "Liquid Phase Epitaxy of Hexagonal Ferrites," *Mat. Res. Bull.* 10 (1975) 1255-1258.
2. M. T. Elliott, "Effects of Lead Incorporation on the Ferromagnetic Resonance Linewidths of Liquid Phase Epitaxial Grown Yttrium Iron Garnet," *AIP Conf. Proc.* 29 (1976) 676-677.

3. M. T. Elliott and H. L. Glass, "Growth-Induced Anisotropy in Yttrium Iron Garnet Films Grown by Liquid Phase Epitaxy," AIP Conf. Proc. 29 (1976) 115-116.
4. H. L. Glass, "Growth of Thick Single-Crystal Layers of Yttrium Iron Garnet by Liquid Phase Epitaxy," J. Crystal Growth 33 (1976) 183-184.
5. H. L. Glass and M. T. Elliott, "Attainment of the Intrinsic FMR Linewidth in Yttrium Iron Garnet Films Grown by Liquid Phase Epitaxy," J. Crystal Growth 34 (1976) 285-288.
6. F. S. Stearns and H. L. Glass, "Liquid Phase Epitaxy of Hexagonal Ferrites and Spinel Ferrites on Non-Magnetic Spinel Substrate," Mat. Res. Bull. 11 (1976) 1319-1326.
7. H. L. Glass, J. H. W. Liaw and M. T. Elliott, "Temperature Stabilization of Ferrimagnetic Resonance Field in Epitaxial YIG by Ga, La Substitution," Mat. Res. Bull. 12 (1977) 735-740.
8. H. L. Glass, "Annealing-Induced Relief of Compressive Misfit Strain in Liquid Phase Epitaxial Yttrium Iron Garnet Films," J. Crystal Growth 40 (1977) 205-213.
9. H. L. Glass and F. S. Stearns, "Growth of Hexagonal Ferrite Films by Liquid Phase Epitaxy," 1977 INTERMAG Conference IEEE Trans. Mag. MAG-13 (1977) 1241-1243
10. H. L. Glass and J. H. W. Liaw, "Growth and Characterization of LPE Hexagonal Ferrites," invited lecture 1977 Conference on Magnetism and Magnetic Materials. J. Appl. Phys. 49 (1978) 1578-1581.
11. H. L. Glass and J. H. W. Liaw, "LPE Growth of Lithium Ferrite on Spinel Substrate Crystals," Mat. Res. Bull. 13 (1978) 353-359.
12. V. S. Speriosu, H. L. Glass and T. Kobayashi, "X-Ray Determination of Strain and Damage Distributions in Ion-Implanted Layers," Applied Physics Letters, to be published.

V. SCIENTIFIC INTERACTIONS

In addition to those presentations which were published in conference proceedings and were included in our list of publications (items 2, 3, 9 and 10), the following papers were presented at conferences.

1. H. L. Glass and M. T. Elliott, "The Effects of Pb Incorporation on the Ferrimagnetic Resonance Properties of Yttrium Iron Garnet Films Grown by Liquid Phase Epitaxy," Third American Conference on Crystal Growth, Stanford University, July 1975.

2. H. L. Glass, M. T. Elliott and D. M. Heinz, "Site Preference in Magnetic Garnet Films Grown by Liquid Phase Epitaxy," Tenth International Congress of Crystallography, Amsterdam, August 1975.
3. H. L. Glass, M. T. Elliott and F. S. Stearns, "LPE Ferrites for Microwave Devices, : American Association for Crystal Growth Western Regional Section, Los Angeles, June 1976.
4. H. L. Glass, "Annealing-Induced Relief of Compressive Misfit Strain in Epitaxial Garnet Films," American Crystallographic Association Winter Meeting, Asilomar, California, February 1977.
5. H. L. Glass and F. S. Stearns, "Liquid Phase Epitaxy of Hexagonal Ferrites from Melts Containing ZnO," Fifth International Conference on Crystal Growth, Boston, July 1977.
6. H. L. Glass, J. H. W. Liaw and F. S. Stearns, "Liquid Phase Epitaxy of Ferrites on Spinel-Structure Substrate Crystals," 4th International Conference on Vapour Growth and Epitaxy.
7. H. L. Glass and V. S. Speriosu, "Double-Crystal Diffraction Profiles from Heteroepitaxial Thin Films: Are the Oscillations Pendellösung?", to be presented to 1979 Annual Meeting of the American Crystallographic Association, University of Hawaii, March 1979.

Our interactions included meetings and discussions with people from a variety of universities, companies and Government laboratories. We have held discussions with our counterparts working on epitaxial microwave ferrites at Westinghouse, University of Texas, MIT, Osaka University, State University of New York and Nippon Electric Company. We have had numerous contacts with DoD laboratory personnel such as J. Adair of AFAL; M. G. Mier of AFML; J. C. Sethares of RADC; A. Tauber, S. Dixon and E. Mariani of ECOM, G. T. Rado, H. Lessoff, C. Vittoria, D. W. Forester, J. Milstein and D. C. Webb of NRL. We have provided samples of our epitaxial ferrite materials to many investigators at universities and at DoD laboratories.

VI. INVENTIONS

During the four year contract period, over a dozen ideas and discoveries were disclosed to our company Patent Department for evaluation. All of these ideas and discoveries were also disclosed to AFOSR and Abstracts of New Technology were submitted in accordance with the terms of the contract.

Thus far, the ideas and discoveries have led to the filing of six patent applications, one of which was subsequently abandoned. Action on the five outstanding applications has not been completed; however, Notices of Allowance have been received from the Patent Office for two of the five. These two applications concern the basic LPE process for hexagonal ferrites and the use of multiple ferrite (YIG) layer structures for signal processing devices.

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REPORT DOCUMENTATION PAGE		READ INSTRUCTIONS BEFORE COMPLETING FORM										
1. REPORT NUMBER AFCR-TR- 79-0195	2. GOVT ACCESSION NO.	3. RECIPIENT'S CATALOG NUMBER										
4. TITLE (and Subtitle) EPITAXIAL GARNETS AND HEXAGONAL FERRITES		5. TYPE OF REPORT & PERIOD COVERED FINAL 01 JAN 75 - 31 DEC 78										
		6. PERFORMING ORG. REPORT NUMBER C79-127/501										
7. AUTHOR(s) H. L. GLASS		8. CONTRACT OR GRANT NUMBER(s) F44620-75-C-0045										
9. PERFORMING ORGANIZATION NAME AND ADDRESS ROCKWELL INTERNATIONAL CORPORATION ELECTRONICS RESEARCH CENTER P. O. BOX 4761 ANAHEIM, CALIFORNIA 92803		10. PROGRAM ELEMENT, PROJECT, TASK AREA & WORK UNIT NUMBERS 61102F/2306B1										
11. CONTROLLING OFFICE NAME AND ADDRESS AIR FORCE OFFICE OF SCIENTIFIC RESEARCH/NE Bldg. 410 Bolling AFB, Washington D.C. 20332		12. REPORT DATE 13 FEB 79										
		13. NUMBER OF PAGES 6										
14. MONITORING AGENCY NAME & ADDRESS (if different from Controlling Office)		15. SECURITY CLASS. (of this report) UNCLASSIFIED										
		15a. DECLASSIFICATION/DOWNGRADING SCHEDULE										
16. DISTRIBUTION STATEMENT (of this Report) Approved for public release; Distribution unlimited.												
17. DISTRIBUTION STATEMENT (of the abstract entered in Block 20, if different from Report)												
18. SUPPLEMENTARY NOTES												
19. KEY WORDS (Continue on reverse side if necessary and identify by block number)												
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20. ABSTRACT (Continue on reverse side if necessary and identify by block number)												
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